LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

UCRL-CONF-231587

# MGA++ Analysis of Low Quantity Samples of U and Pu on an Extended-rage Gamma-ray Detector

T.-F. Wang, G. P. Russ, R. W. Williams

June 7, 2007

48th INMM Annual Meeting
Tucson, AZ, United States
July 8, 2007 through July 12, 2007

# MGA++ analysis of low quantity samples of U and Pu on an extended-rage gamma-ray detector

Tzu-Fang Wang, G. Price Russ, and Ross W. Williams
Lawrence Livermore National Laboratory, Livermore, CA 94550

## Introduction:

The IAEA has expressed a need for improved determination of gamma emitting nuclides in environmental samples collected during inspections of nuclear facilities and to use the MGA++ to determine U and Pu concentrations and isotopic compositions when those elements are present in relatively high concentrations. We are addressing the IAEA needs by evaluating the applicability of extended-range germanium detectors (ERG). In this paper we used 1g U isotopic standards and 100ug Pu liquid standards 1) to determine the performance of MGA++ on this special detector and 2) to estimate the amount of U and Pu necessary in the sample for determination of the isotopics via MGA++ within reasonable accuracy for a week of counting time using this ERG detector.

## MGA++:

The gamma-ray multi-group analysis code MGA++ [1,2,3] developed at Lawrence Livermore National Laboratory has been widely used in the area of gamma-ray non-destructive plutonium/uranium assay. This plutonium/uranium isotopic analysis code de-convolutes the complicated, 100-keV x ray and gamma ray region to obtain the ratio of Pu and U isotopes. Calibration of the detector efficiency is not required, but is determined intrinsically from the measured spectra.

## The Detector:

We have selected an. ORTEC PROFILE FX-Series GEM Coaxial P-type HPGe Gamma-Ray detector (Model GEM-FX8530) for this study. The detector has a diameter of 85 mm and is 30 mm in length, Its resolution is about 700eV@ 122 keV and 1.9keV@ 1.33 MeV with a nominal relative efficiency of 50%. The reasons for selecting this detector were: 1) Maximum absolute counting efficiency over a broad energy range with minimum uncertainties for “close-in geometry” counting; 2) The ability to use a known geometry close to the end cap; 3) Large diameter to minimize the errors due to unknown source distribution of the swipes and 4) High resolution at low and high energy. We have also selected low-background cryostat construction for background minimization.

## Experimental Setup

Four uranium oxide standards and one plutonium liquid standard were used in this study. The uranium oxide standards are NBL CRM U005 (0.5%), CRM U010 (0.99%), CRM U500(49.4%), and CRM U750 (75.2%) and the plutonium liquid standard is NBL CRM126. The percentage numbers in parentheses are the certified weight percentage of 235U/U. We have not tried the medium (5-20%) uranium enrichments because we believe that if both depleted-natural and high enriched measurements are in good agreement with the declared values, the medium-enriched will have good agreement also. This is because the gamma-ray intensity of the 235U and 238U daughters in the 100-keV region are roughly equally intense for the medium enriched uranium. Signals were collected using ORTEC DSPEC$^{plus}$ spectrum analyzers at 0.075 keV/channel for 16k

channels with a shaping time of 4μS. Uranium spectra were collected for 600 second intervals and continuous dumping for 10 hours. For the plutonium liquid standards data were collected at a 2 hr intervals for 80 hours.

**MGA++ analysis and results:**

We have determined that anomalies recently found in the MGA++ -U235 code[4]) were due to the special background determination algorithm used in the code. For this work, we used the in house MGA4U[5]) code which employed LLNL commonly used background subtraction for the following analysis:

*1) CRM U005 standard:*

Figure 1 shows results from MGA4U deconvolution. From the limitation in the deconvolution algorithm combined with the detector resolution (700eV@ 120 keV as opposed to 600eV@120keV as recommended by the MGA4U), we have slight positive bias in the results for this depleted uranium standard. It is also estimated that our sensitivity for a week of counting time would be about 1mg with 5% (two sigma) of uncertainty in the isotopic ratio. The corresponding % uncertainties shown in Figure 2 demonstrate that MGA4U employed statistical uncertainty in the intensity of the peaks.

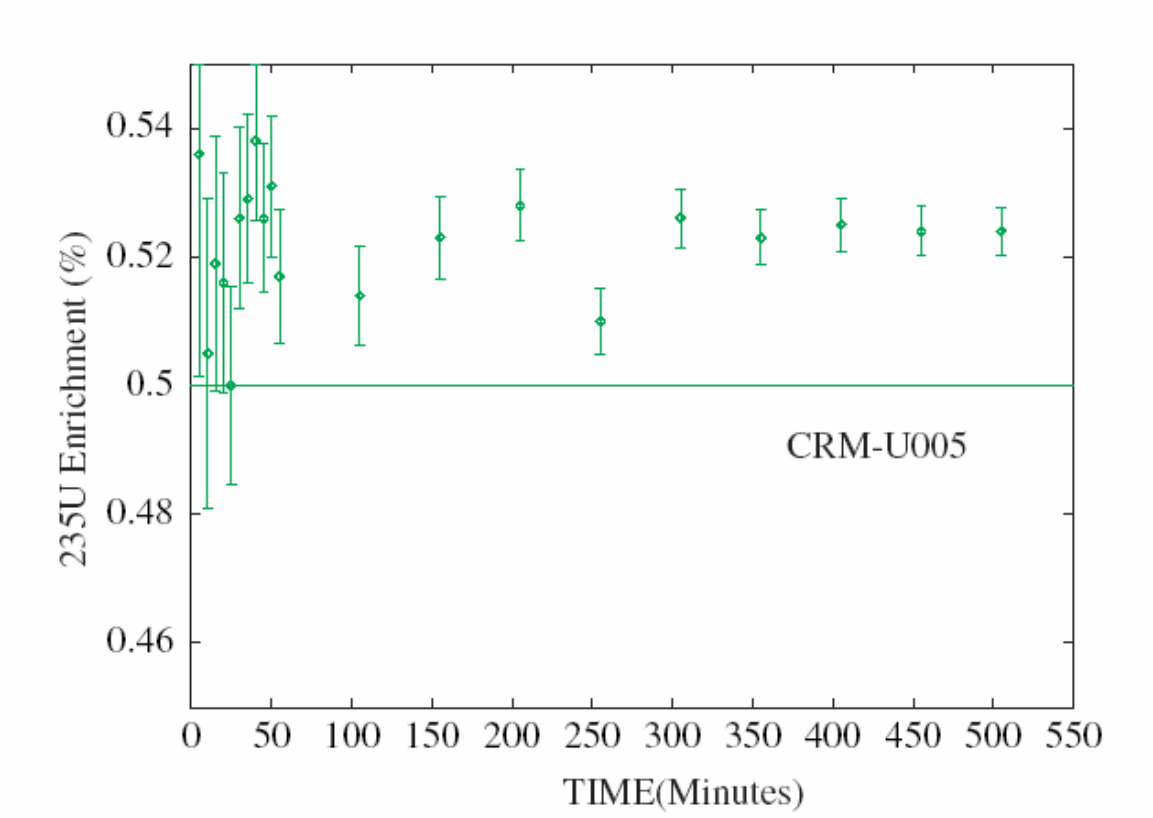


Figure 1 The 235U in weight % from MGA4U deconvolution w.r.t. time. The horizontal line represents the certified value.

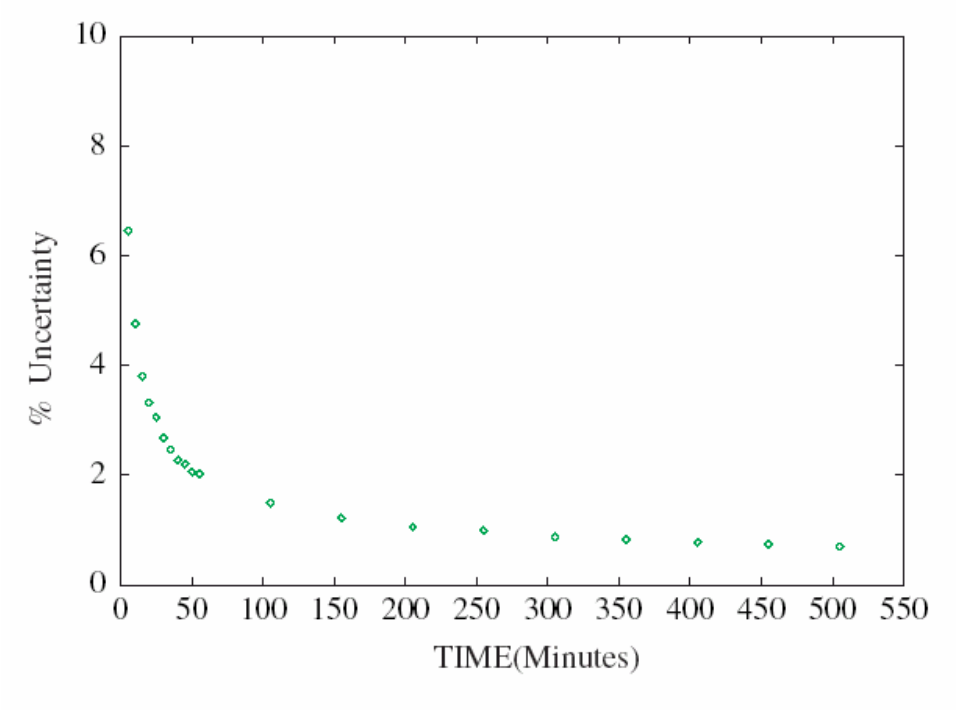


Figure 2. The % uncertainty w.r.t. time.

*2) CRM U010 standard:*
Figure 3 shows the results. The expected sensitivity (as described above) is about 0.5mg

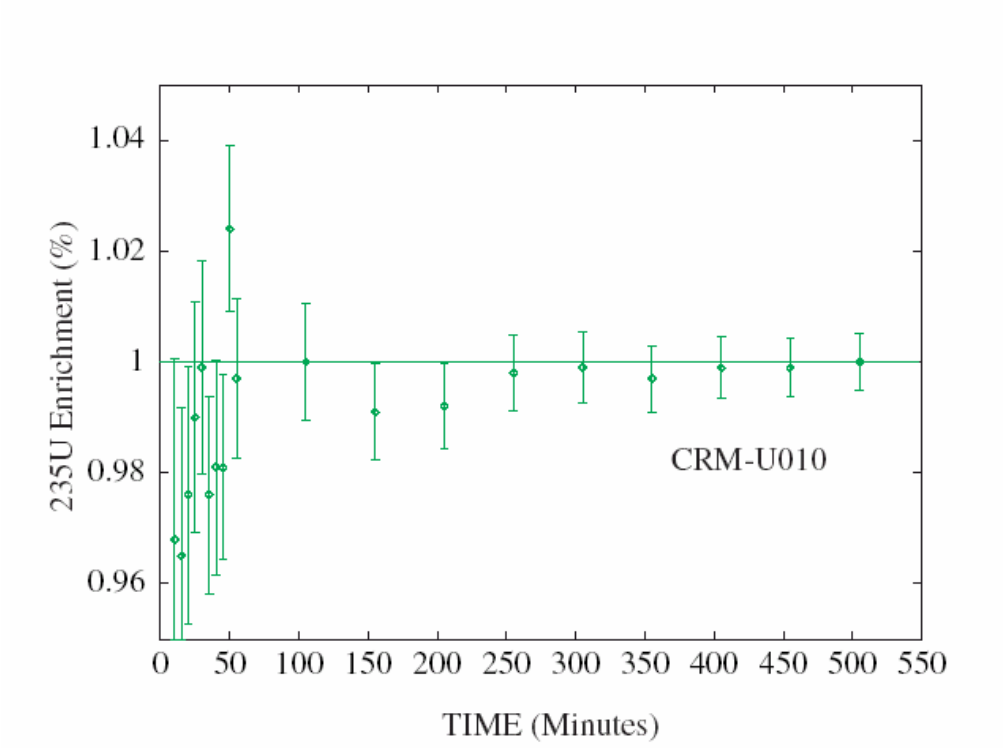


Figure 3 The 235U in weight % from MGA4U deconvolution w.r.t. time. The horizontal line represents the certified value.

*3) CRM U500 standard:*
Figure 4 shows the results. The results show a slight negative bias, particularly with increased counting time, with the MGA4U/Detector combination. The expected sensitivity (as described above) is about 0.5mg.

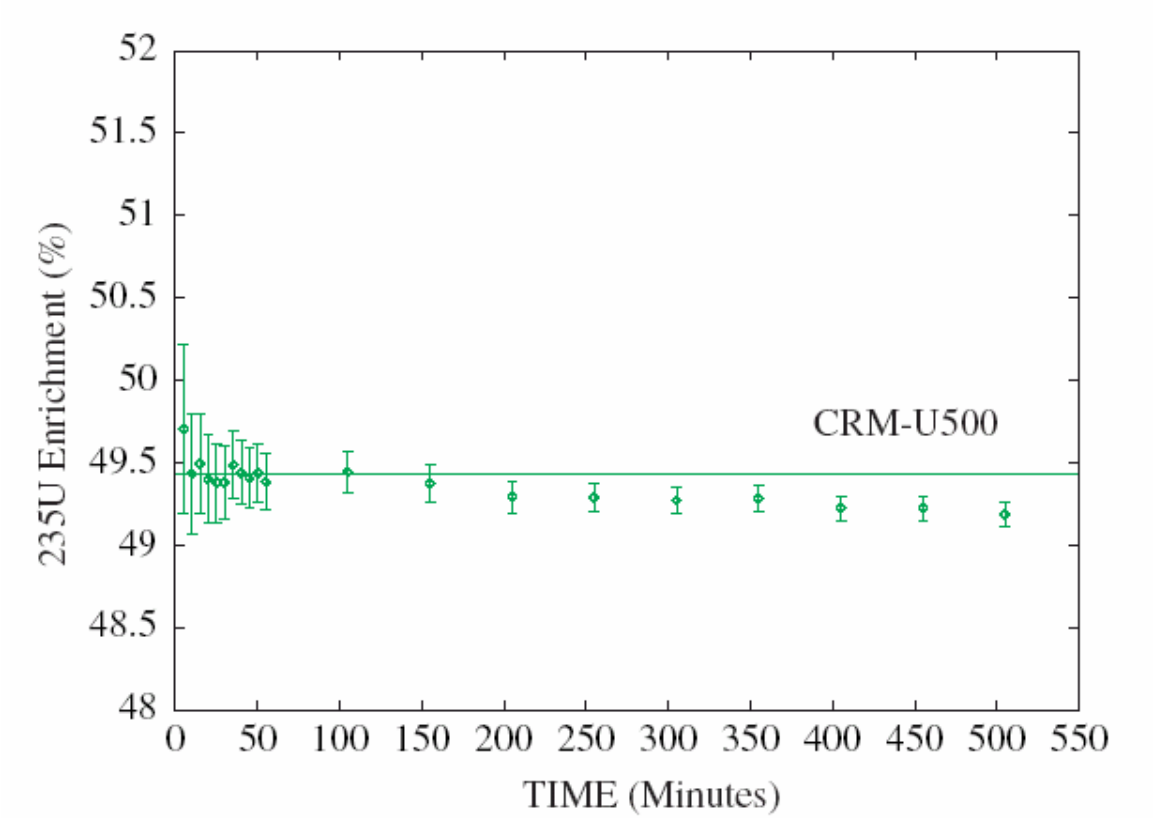


Figure 4 The 235U in weight % from MGA4U deconvolution w.r.t. time. The horizontal line represents the certified value.

*4) CRM U750 standard:*
Figure 5 shows the results. For short count time, the results show a positive bias with the MGA4U/Detector combination. The expected sensitivity (as described above) is about 2.5mg.

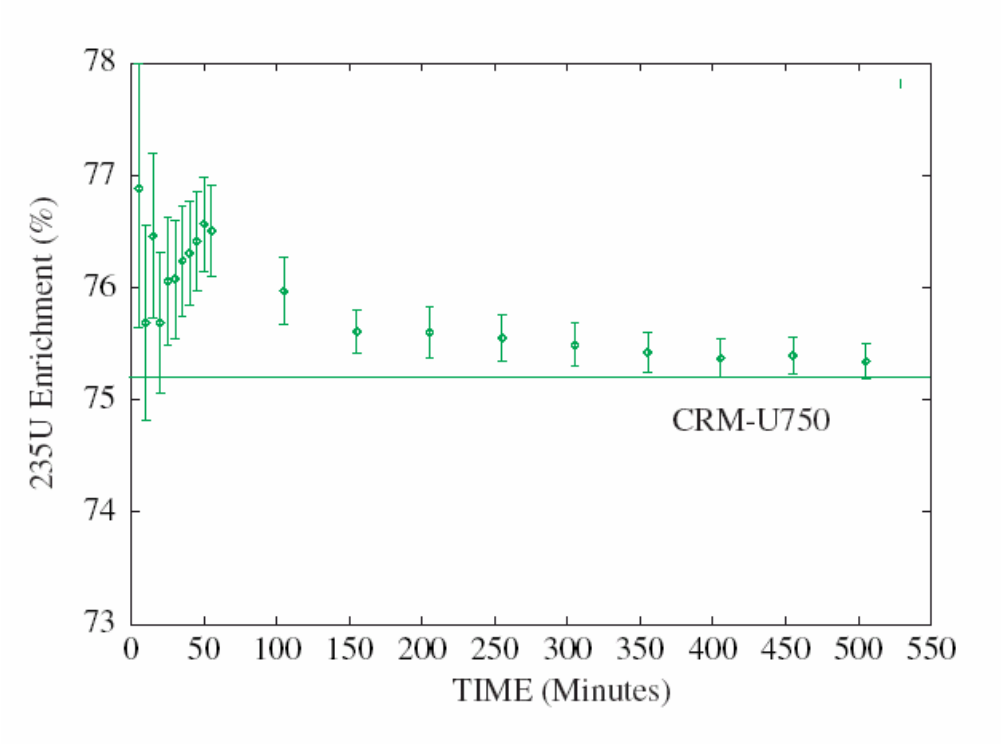


Figure 4 The 235U in weight % from MGA4U deconvolution w.r.t. time. The horizontal line represents the certified value.

*5) CRM 126 plutonium standard:*
Table 1 shows the MGA++ deconvolution results for the CRM 126 plutonium standard. The MGA++ failed to deconvolute the data till about 60 Hr of collecting time. Our estimated sensitivity is about 100ug

| | Pu-238 (%) | Pu-239 (%) | Pu-240 (%) | Pu-241 (%) | Pu-242 (%) |
|---|---|---|---|---|---|
| Declared | 0.0122 | 93.886 | 5.915 | 0.149 | 0.036 |
| Measured | 0.0129 | 93.856 | 5.911 | 0.190 | 0.030 (calc.) |
| % Uncertainty | 11.67 | 0.04 | 1.97 | 5.63 | 10 |

Table I Results from MGA++, the Pu-242 value is derived from an empirical[3)] formula.

**Conclusions:**
The automatic efficiency calibration algorithms work very well in the new ERG detector test. The "close" gamma-ray energy (i.e. using nearby-energy of characteristic gamma-rays) isotopic deconvoluation methodology employed in the MGA++ also minimized the detector efficiency effects in the isotopics determinations. The time dependent uranium isotopic ratio observed above is due to the overall efficiency determination using the available gamma-ray information, especially in fitting of the Kβ region. This task is the first step to demonstrate that ERG/MGA++ performed adequately with a wide range of uranium samples in the mg range and low-burnup 100ug plutonium samples. We will perform more measurements using "scattered" samples (to simulate radioactivity on a filter paper) to further evaluate the ability of the ERG/MGA++ combination.

**Acknowledgements**:
We thank Jim Brunk, Cindy Conrado, and Robert Haslett in the setting up of the ERG detector. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

**References:**

1) W. E. Parker, T.F. Wang, D. Clark, W. M. Buckley, W. Romine and W. D. Ruhter, *Plutonium and Uranium Isotopic Analysis: Recent Developments of the MGA++ Code Suite,* Proceedings of the Sixth International Meeting on Facilities Operations - Safeguards Interface, pp. 192 - 197, American Nuclear Society, Jackson Hole, Wyoming, September 1999.
2) R. Keyser, T. Twomey, S. Haywood, W. E. Parker, T.F. Wang, D. Clark, K. Raschke, W. Romine, W. Buckley and W. Ruhter, *Recent Developments in the MGA++ Codes,* ESARDA Conference, Seville, Spain, May 1999.
3) R. Gunnink, W.D. Ruhter, *MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances*, Volume I and II, Lawrence Livermore National Laboratory, Livermore, CA., UCRL-LR-103220, (1990)
4) See Dr. Alexander Solodov et al., "Achievements on Nondestructive Assay Isotopic Enrichment Measurement Systems under the USDOE/NNSA-ABACC Safeguards Cooperation Agreement" in this INMM (2007).
5)T. F. Wang, W. D. Ruhter, R. G. Lanier, *Re-Measured Uranium Branching Ratios and thier Impact on Removing Biases from MGAU Analyses,* Proceedings of the Sixth International Meeting on Facilities Operations - Safeguards Interface, pp. 220 - 223, American Nuclear Society, Jackson Hole, Wyoming, September 1999.